Hybridization-induced superconductivity from the electron repulsion on a tetramer lattice having a disconnected Fermi surface

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Plaquette lattices with each unit cell containing multiple atoms are good candidates for disconnected Fermi surfaces, which are shown by Kuroki and Arita to be favorable for spin-flucutation mediated superconductivity from electron repulsion. Here we find an interesting example in a tetramer lattice where the structure within each unit cell dominates the nodal structure of the gap function. We trace its reason to the way in which a Cooper pair is formed across the hybridized molecular orbitals, where we still end up with a T_c much higher than usual.

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The discovery of high-T_c superconductors [1] has kicked off renewed interests in electronic mechanisms for superconductivity. In the course of studies it is becoming increasingly clear that superconductivity can arise from repulsive interaction between electrons. The essence there is electrons interact by exchanging spin fluctuations, which can effectively act as an attraction in the gap equation, if we consider anisotropic pairing with nodes in the gap function.

For the Hubbard model, this was first suggested from early calculations by Scalapino et al. [5]. Subsequently a quantum Monte Carlo calculation [6] indeed indicated an enhancement of the pairing correlation with d-wave symmetry in the repulsive Hubbard model. The superconducting critical temperature (T_c) has been estimated with the fluctuation-exchange approximation (FLEX), a kind of renormalized random phase approximation. [2–4]

One remarkable point is $T_c \sim O(0.01t)$, esitmated for the repulsive Hubbard model in the two-dimensional (2D) square lattice, is *two orders of magnitudes smaller than* the starting electronic energy (i.e., the hopping integral t), although this gives the right order for the curates' T_c. Arita et al. then looked at various lattices (square, trianglar, fcc, bcc, etc) in search of the most favorable case with a FLEX analysis [7]. The best case, as far as these ordinary lattices are concerned, turns out to be the 2D square lattice, so $T_c < O(0.01t)$ remains. As discussed in Ref. [8], there are good reasons why T_c is so low, where an important one is the presence of nodes in the superconducting gap function greatly reduces T_c: While the main pair-scattering, across which the gap function has opposite signs to make the effective interaction attractive, some of the pair scatterings around the nodes have negative contributions to the effective attraction by connecting k-points on which the gap has the same sign.

So a next important avenue to explore is: can we improve the situation by going over to multiband systems. Kuroki and Arita [8] have shown that this is indeed the case if we have *disconnected Fermi surfaces*. In this case T_c is dramatically enhanced, because the sign change in the gap function can avoid the Fermi pockets, where all the pair-scattering processes contribute positively. [8–10] This has been numerically shown to be the case for the triangular lattice (for spin-triplet pairing) [11] and a squre lattice with a period-doubling [8], where T_c as estimated with FLEX is as high as $O(0.1t)$.

To be more precise, the key ingredinents are: (a) when the Fermi surface is nested, the spin susceptibility $\chi(\boldsymbol{q},\omega)$ has a peak, with a width $\Delta\boldsymbol{Q}$. (b) When a multiband system with a disconnected Fermi surface has an inter-pocket nesting (i.e., strong inter-pocket pair scattering and weak intra-pocket one) the gap function has the same sign (s-wave symmetry) within each pocket, and the nodal lines can happily run in between the pockets.

Here a natural question is: to prepare multiband systems systematically, we can consider lattices comprising some units, or "plaquette lattices". This is indeed considered by Kimura et al. [9] who conceived an idea of actually fabricating the structure from arrays of quatum dots. One example is the inset of Fig. 1, where the unit is a square lattice of squres, where the intra-plaquette transfer is stronger than the inter-plaquette one. Now, an important question is: how the structure of the superconducting gap function having higher T_c's is determined and whether the s-wave symmetry on each pocket is compulsory.

In this paper, we study the repulsive Hubbard model on a square lattice of diamonds, where a diamond is a four-site unit rotated by 45 degrees (Fig. 1). One motivation is that this structure is reminiscent of the in-plane lattice of B and C in CaB_2C_2 [12], so should not be too unrealistic, although we are not claiming to consider this

particular material. We have solved Eliashberg's equation [13] for the Hubbard model with the FLEX and obtained a T_c that is considerably higher than that of the square lattice. More importantly, however, the present model, despite of its disconnected Fermi surface, has sign changes in the gap function within each pocket, so a different mechanism should be at work. We can trace back its reason in *real space* that the singlet pairing here results from a hybridization of two *molecular orbitals* in each plaquette.

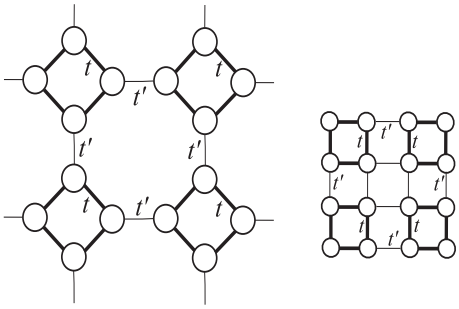

FIG. 1. The lattice considered in this paper. The inset shows the plaquette lattice in Ref. [9].

So a message of this work is that, in order to search for higher T_c's, we should look not only at the Fermi surface but also at the atomic structure in real space.

We have used the four-band version of FLEX, [9,10,14] where the Green's function G, the spin susceptibility χ, the self-energy Σ, and the superconducting gap function ϕ are 4×4 matrices, e.g., $G_{lm}(\boldsymbol{k}, i\varepsilon_n)$, where l, m specify the four sites in a unit cell. We can go from the site indeces over to band indeces with a unitary transformation. For the spin susceptibility, we concentrate on its largest eigenvalue denoted as χ. From Green's function and the spin susceptibility we obtain the superconducting gap function (with spin-singlet pairing assumed) and T_c by solving the linearized Eliashberg's equation [13]. In our analysis, we take 32×32 k-point meshes and up to 4096 Matsubara frequencies, where the numerical results are sufficiently converged. The on-site Hubbard repulsion is taken to be a typical strong-correlation value, $U/W = 7/6$, where W is the band width. The band filling (=number of electrons/number of sites) is taken to be a value $n = 0.85$, which is close, but not too close, to the half-filling where the Mott transition and antiferromagnetic order are expected. In the below we have confirmed that when the eigevalue of Eliashberg's equation becomes unity prior to the divergence of the spin susceptibility.

The FLEX result for the spin susceptibility, Green's function, and the superconducting gap function are shown in Fig. 2 for $t' = 0.6$ ($t = 1.2$) with $T = 0.06$. Ridges in the Green's function delineate the Fermi surface. Fermi surface comprises two bands (second and third from the bottom), which we call band A and band B. We can see that each band has a squre-like pocket for the Fermi surface, which is reminiscent of the Fermi surface of Sr_2RuO_4, with a good reason as we shall see. On the other hand we notice that the spin susceptibility has broad peaks around (π, π). Quite unexpectedly, the gap function has a strange structure in such as way that its amplitude on the Fermi surface is peaked at the *corners* rather than along the edges.

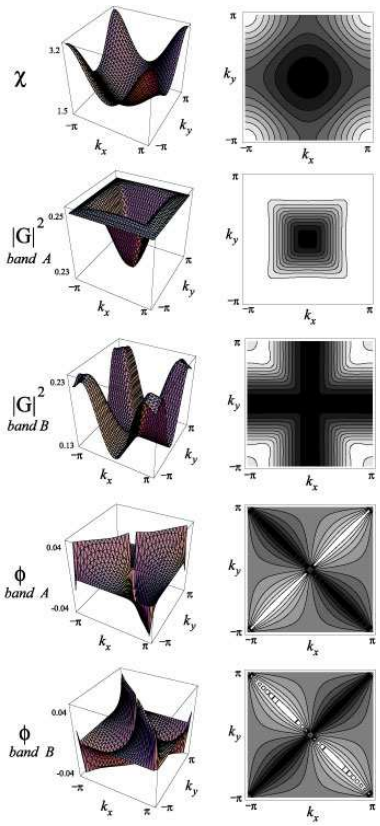

FIG. 2. Spin susceptibility (χ), the absolute value of the Green's function squared ($|G|^2$), and the superconducting gap function Φ against k_x, k_y for the lowest Matsubara frequency for $U = 7$, $n = 0.85$, $t = 1.2$, $t' = 0.6$, and $T = 0.06$.

Figure 3 shows T_c as a function of t. [15] A plaquette lattice is characterized by the intra-plaquette transfer t and inter-plaquette one t'. To facilitate comparison, we have fixed the single-particle bandwidth $W = 2(2|t|+|t'|)$ to 6, which is the value of W when $t = t' = 1$. Interestingly, T_c is larger for smaller $|t'|$, which is in accord with our expectation that the case of plaquette lattice (tetramirization in the present case) is favorable. T_c saturates to a value $T_c = 0.06 = 0.01W$, which is two times greater than that ($< 0.03t$) for the square lattice.

The enhancement of T_c in smaller $|t'|$ can be understood by the nesting between the disconnected Fermi surface (Fig. 4). At half filling ($n = 1$) the Fermi surface is perfectly nested regardless of the value of $|t'|$. When less than half-filled, the nesting in the band depicted by $\boldsymbol{Q}$ in

Fig. 3 degrades but not so much for smaller $|t'|$. Because the effective attraction here is mediated by the antiferromagnetic spin fluctuation (with a large $|Q|$), T_c is larger for better nesting, i.e., for smaller $|t'| \propto$ the warping of the quasi-1D Fermi surface. This t'-dependence of T_c can be understood also in real space, where the intra-cell spin singlet should be more robustly formed for smaller $|t'|$.

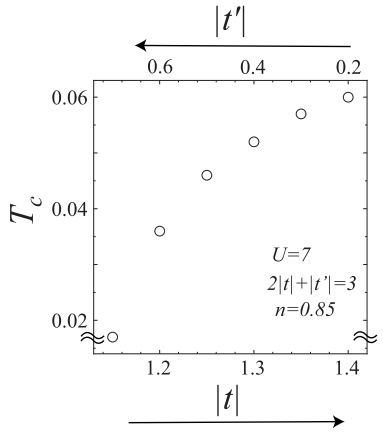

FIG. 3. T_c plotted as a function of t for $U = 7$ and $n = 0.85$ in units where the band width $2(2t + t') = 6$.

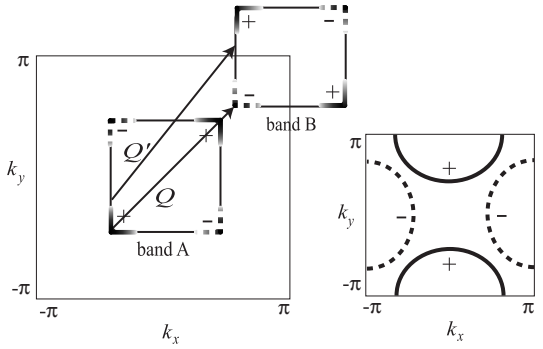

FIG. 4. Fermi surfaces of the lattice depicted in Fig.1. The solid (dashed) curves represent positive (negative) values of the gap function on the Fermi surface. The arrows represent typical momentum transfer in pair scattering processes, where $\boldsymbol{Q}$ corresponds to the wave vector at which the spin susceptibility is peaked. The solid (dashed) curves in the inset represents the positive (negative) values of the gap function in a plaquette lattice in Ref. [9].

We can explain the structures of the strange gap function in terms of molecular orbitals (Fig. 5a) and the pairing (Fig. 5b) in real space. As mentioned, the Fermi surface reminds us of the (α and β) bands in SrRuO, which consists of two sets of quasi-1D array of orbitals. In the present case the quasi-1D band structure arises from the molecular orbitals in the plaquette. The molecular orbitals that are relevant to bands A and B have p_x- and p_y- symmetries (Fig. 5a), so that we do have two sets of 1D arrays of orbitals. Due to the hybridization of the two orbtials, the two sets of quasi-1D Fermi surface (two, orthogonal sets of parallel lines) anticross, and we end up with two square-shaped pockets.

Now the interesting point is, the Cooper pair, arising from antiferromagnetic fluctuations, is formed across the adjacent sites (ellipses in Fig. 5b), that is *across* the two molecular orbitals. This implies that the gap function on the Fermi surface is large only around the positions at which the two (p_x and p_y) bands hybridize, i.e., around the anticrossing points. The pairings depicted in Fig. 5b as solid ellipses and dashed ellipses have opposite signs, so that the symmetry is d_{xy}. We have thus a curious case of an inter-band pairing (a pairing formed by inter-band pair scatterings). We can in fact confirm that the pairing is interband and d_{xy} by looking at the sign of the gap function. The ellipses is Fig. 5 are represented by $\langle C_{A(B)\uparrow} C_{A(B)\downarrow} \rangle$, where $C_{A(B)\sigma} \equiv C_{1\sigma} - C_{2(4)\sigma} + C_{4(2)\sigma} - C_{3\sigma}$, where $C_{i\sigma}$ annihilates an electron with spin σ at site i as numbered in Fig.5. When the pairing is formed across adjacent sites we can show $\langle C_{A\uparrow} C_{A\downarrow} \rangle = -\langle C_{B\uparrow} C_{B\downarrow} \rangle$ (opposite signs across the bands) with an odd parity (d_{xy}) against $k_x \leftrightarrow -k_x$ or $k_y \leftrightarrow -k_y$.

This explains why the gap function changes sign within each pocket. Hence the s-wave symmetry on each pocket is not a necessary condition for a high T_c. Even when there are intra-pocket sign changes, the disconnectivity of the Fermi surface plays an important role, since the *inter-pocket* sign change associated with $\boldsymbol{Q}$ does exploit this in avoiding intersecting the Fermi surface.

One might then wonder why the intra-pocket sign changes do not reduce T_c, since the pair-scattering processes depicted by $\boldsymbol{Q}'$ in Fig. 4 connecting the k-points across which the gap function has the same sign would normally reduce T_c. In the present case, however, we have an ingeneous situation where the gap function is strongly peaked at the position (corners in this example) where hybridization occurs, so that the $\boldsymbol{Q}'$ processes, being off the peak, have little effect.

How is the present model compared with the lattice considered in Ref. [9], where the plaquette is a four-membered ring in either case. Here the consideration in real space comes in handy. The latter case is a 2D system rather than quasi-1D, where the pockets arise due to a band folding due to the tetramerization,. The pairing in real space is formed within each molecular orbital (Fig. 6) unlike the present case, so the superconducting gap function has an s-wave symmetry on each Fermi surface pocket (but opposite signs across the two pockets with the nesting $\boldsymbol{Q} \approx 0$ due to the band folding (inset of Fig. 4). Despite of this distinction, the resulting T_c's are similar between the two cases.

Another interesting comparison is with Sr_2RuO_4 [16,17], which also suggests an importance of the real space structure. Although the Fermi surface is quasi-1D in Sr_2RuO_4 as well, the oxide has strong pieces of evidence for triplet pairing, and even when one considers the spin-singlet pairing, T_c (for the α, β bands for this material) estimated with the FLEX is very small [17]. Due to the quasi-one dimensionality the spin susceptibility $\chi(\boldsymbol{q}, \omega)$ has linear ridges in k-space. As a result, the pair scatterings have large contributions all over the ridges, so that the (extended) s-wave gap function involves unfavorable pair scatterings across which the gap has the same sign, resulting in the reduced T_c. This makes the spin-triplet, p-wave pairing more favorable, for which the resulting gap function has large amplitudes all over the Fermi surface, except for dips at the corners . By contrast, the present case has a built-in antiferromagnetic structure within the unit cell in real space, so that the spin susceptibility has a well-defined peak around (π, π). This makes a specific $\boldsymbol{Q}$ to be relevant in the pair scattering, which makes the gap function peaked at specific points, which in turn gives rise to an enhancement in T_c.

To summarize, we should question not only the structure of the Fermi surface but also the structure of the molecular orbitals and the superconducting pairings in real space in understanding the superconductivity from the electron repulsion. One interesting tendency is that high T_c's are found in plaquette systems where unit cells having multiple sites are connected with a relatively small inter-plaquette hopping [8–10], which also suggests the importance of the real space picture. In fact, the relation between real-space and the momentum-space pictures has been discussed for systems consisting of dimers recently. [10] It is an interesting future problem to see how the real-space and the momentum-space pictures are related with each other for wider class of systems in realizing high T_c's.

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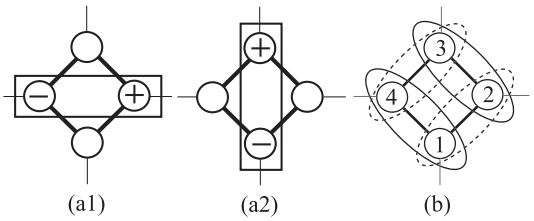

FIG. 5. (a) Intra-plaquette molecular orbitals with p_x (a1) and p_y (a2) symmetries. (b) Cooper pairs with solid and dashed ellipses indicating pairing amplitudes with opposite signs.

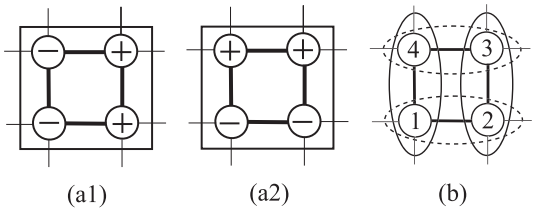

FIG. 6. (a) Intra-plaquette molecular orbitals with p_x(a1) and p_y(a2) symmetries, and (b) Cooper pairs for the lattice considered in Ref. [9].

[1] J.G. Bednorz and K.A. Müller, Z. Phys. B **64**, 189 (1986).
[2] N.E. Bickers, D.J. Scalapino, and S.R. White, Phys. Rev. Lett. **62**, 961 (1989).
[3] S. Grabowski, M. Langer, J. Schmalian, and K.H. Bennemann, Europhys. Lett. **34**, 219 (1996).
[4] T. Dahm and L. Tewordt, Phys. Rev. B **52**, 1297 (1995).
[5] See, e.g., D.J. Scalapino, E. Loh, and J.E. Hirsch, Phys. Rev. B **35**, 6694 (1987); S.R. White, D.J. Scalapino, R.L. Sugar, N.E. Bickers, and R.T. Scalettar, Phys. Rev. B **39**, 839 (1989).
[6] K. Kuroki and H. Aoki, Phys. Rev. B **56**, R14287 (1997).
[7] R. Arita, K. Kuroki, and H. Aoki, J. Phys. Soc. Jpn. **69**, 1181 (2000).
[8] K. Kuroki and R. Arita, Phys. Rev. B **64**, 024501 (2001).
[9] T. Kimura, H. Tamura, K. Kuroki, K. Shiraishi, H. Takayanagi, and R. Arita, unpublished (cond-mat/0204218).
[10] K. Kuroki, T. Kimura, and R. Arita, unpublished (cond-mat/0206204).
[11] K. Kuroki and R. Arita, Phys. Rev. B **63**, 174507 (2001).
[12] B. Albert and K. Schmitt, Inorg. Chem. **38**, 6159 (1999).
[13] G.M. Eliashberg, Zh. Eksp. Teor. Fiz. **38**, 996 (1960) [Soviet Phys. JETP **11**, 696 (1960)].
[14] K. Kuroki, T. Kimura, R. Arita, Y. Tanaka, and Y. Matsuda, Phys. Rev. B **65**, 100516 (2002).
[15] T_c depends only on the absolute values of t or t'. The invariance under $t \to -t$ and/or $t' \to -t'$ can be shown with a gauge transformation, $C_{1(3)} \to -C_{1(3)}$ and $C_{2(4)} \to -C_{2(4)}$ for $t \to -t$, and $C_n \to (-1)^{x+y} C_n$ $(n = 1, 2, 3, 4)$ for $t' \to -t'$, where x and y are the position of the unit cell measured in units of the cell size.
[16] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J.G. Bednorz, and F. Lichtenberg, Nature (London) **372**, 532 (1994).
[17] K. Kuroki, M. Ogata, R. Arita, and H. Aoki, Phys. Rev. B **63**, 060506(R) (2001).